P-75

The effect of mechanical activation on the relaxor properties of $PbMg_{1/3}Nb_{2/3}O_3$ - $PbFe_{1/2}Nb_{1/2}O_3$ solid solution ceramics

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Recently we have found out that the high-energy mechanical activation of the starting oxides reduces dramatically the frequency shift ΔT and increases by about 20 K the temperature T_m of the dielectric permittivity maximum for ceramics of a classical ferroelectric-relaxor $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) sintered from the mixture of these oxides [1]. Similar changes of ΔT and T_m were predicted previously, basing on the first-principle calculations, for PMN with an increased degree of a short-range ordering of Mg^{2+} and Nb^{5+} cations [2]. However, as we used for mechanical activation a high-energy planetary-centrifugal mill AGO-2 with both jars and balls made of the stainless steel, one could expect that a small amount of iron from a milling media incorporates into the processed powder and the $(1-x)PbMg_{1/3}Nb_{2/3}O_3$ - $xPbFe_{1/2}Nb_{1/2}O_3$ (PMN-xPFN) solid solution is formed during sintering. The formation of such solid solution, in principle, could explain the observed increase of T_m as well as a smaller unit cell parameter value (4.037 Å instead of ≈4.05 Å for a usual PMN ceramics) observed for PMN ceramics studied in [1]. The scope of the present work was to obtain several PMN-PFN solid solution compositions by both the usual solid-state synthesis and by high-energy mechanochemical synthesis and compare their relaxor properties and the values of the unit cell parameters.

Energy dispersive X-ray analysis (EDXA) has shown that the PMN samples obtained using high-energy mechanochemical synthesis contain about 1 wt.% of iron. Raman studies revealed that an additional band appears in the spectrum of such ceramics at 700 cm-1. Similar band is present in the Raman spectrum of PFN and it corresponds to the Fe-O stretching mode [3]. Thus at least a part of iron incorporates into the lattice of PMN. If one assumes that all this iron incorporates into the crystal lattice of PMN, the 0.8PMN-0.2PFN solid solution composition would be formed, which contains some MgO excess. Our studies have shown that for the PMN-PFN compositions from the $0 \le x \le 0.2$ range both the unit cell parameter and the ΔT values are smaller when ceramics is obtained using high-energy mechanochemical synthesis. On the other hand Tm values were very similar for the ceramics of the same composition obtained by both methods. Thus, one may conclude that the increase of Tm in the PMN ceramics obtained using high-energy mechanochemical synthesis may be ascribed, at least partially, to the formation of the PMN-PFN solid solution. However the main origin of the unit cell parameter and ΔT changes observed for such ceramics seems to be not the incorporation of iron from the milling media into the lattice, but rather the effect of high-energy mechanical activation.

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